# Miniband dispersion and excitonic effects on the optical spectra of GaAs/$Al_xGa_{1-x}As$ superlattices

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We have studied the luminescence excitation properties of a series of GaAs/$Al_xGa_{1-x}As$ superlattices under magnetic field. The periods of the samples are such that the miniband widths for electrons range between 0 and 30 meV. New structures, related to the miniband dispersion, are observed at zero magnetic field, some of which are excitonic due to their behavior under magnetic field. Comparing experimental spectra and theoretical calculations, we discuss the possible attribution of these structures to saddle-point excitons.

Coulomb effects lead to drastic changes in the absorption spectra of semiconductors. The most extensively studied is the formation of stable excitons which bring sharp resonances in the optical spectra.[1] Other effects such as Sommerfeld enhancement[2] or excitonic enhancement around the Fermi energy[3,4] are also observed and quite well understood theoretically.

The expansion of the optical transition energy in a Taylor series around one of the critical points of the band structure leads, in a first approximation, to a parabolic development with three reduced-mass parameters. Four types of critical points can be found, labeled $M_j$ according to the number $j$ of negative mass components. $M_1$ critical points are called saddle points due to the shape of the band structure around them. Coulomb interaction between electrons and holes associated with the saddle-point states gives rise to exciton resonances below the energy of the saddle point. In bulk materials, the existence and behavior of saddle-point excitons has been the subject of numerous studies, theoretical[5] as well as experimental.[6] In alkali halides, for example, such excitons are observed as sharp structures in resonance with the band-to-band transitions.[6]

Superlattices (SL's) represent a very interesting case where the zone folding in the growth direction $z$ of the band structure keeps an $M_0$ singularity at the center of the Brillouin minizone (BZ) ($k_z=0$, where $k_z$ is the wave vector along the direction $z$), and leads to the formation of saddle points at the BZ edges ($k_z=\pm\pi/d$, where $d$ is the SL period). By varying $d$, the miniband width can be tailored from less than 1 meV to more than 200 meV. Therefore, it can be made smaller as well as larger than the exciton binding energy.

In this paper, we describe the photoluminescence excitation (PLE) results obtained on a series of SL's with different periods. We compare our experimental results to theoretical calculations,[7] and obtain a very good agreement. The new structures observed in the PLE spectra of the SL's are found to derive from the existence of the miniband structure and of $M_0$ and $M_1$ singularities in the density of states. Their excitonic character is confirmed by the experiments under a magnetic field.

Our GaAs/$Al_xGa_{1-x}As$ samples are grown by molecular beam epitaxy. Precise determination of the SL parameters is obtained by x-ray diffraction:[8] the result of this determination labels the SL ($a/b$ means well width $a$ and barrier width $b$ in Å). The growth conditions have been optimized for short-period structures, and high quality is achieved, characterized by a very good flatness of the interfaces, and a good reproducibility of the layer thicknesses.[9,10]

Magnetoexcitation of the luminescence is performed in a superconducting magnet up to fields of 10 T. In order to obtain the PLE spectra, we set the monochromator on the heavy-hole exciton peak and scan the excitation energy of the krypton-pumped LD700 dye laser (with two possible incident polarizations, $\sigma^+$ or $\sigma^-$).

PLE spectra of superlattices show distinct behavior from quantum-well spectra at zero magnetic field already. Some of these differences have been described elsewhere and arise from the increase in three-dimensional character of the structure as the period decreases[11,12] (see in Ref. 13 that the shape photoreflectance spectra is also affected). When the SL period decreases, the spectrum gradually changes from the shape typical of multiple quantum wells (MQW's) to a shape similar to what is observed in bulk GaAs layers: as an example, the heavy-hole ($HH_0$) and light-hole ($LH_0$) excitonic lines eventually merge into one peak[14] (the index 0 stands for transitions associated with the extremum in $k_z=0$).

The new effect that we study here is depicted in Fig. 1 (each experimental spectrum is above the corresponding theoretical one). Going from the lower curve to the upper one corresponds mainly to a change in the barrier thickness. As a consequence, the miniband widths increase from 0 (MQW case) to $\sim 30$ meV for electrons, 1 meV for HH's, and $\sim 26$ meV for LH's. Sample (52 Å)/(140 Å) exhibits the usual PLE spectrum of a MQW sample, with two excitonic resonance peaks ($HH_0$ and $LH_0$) and two shoulders corresponding to the onset of band-to-band transitions (with the contribution of the excited states of the corresponding excitons). When the SL period decreases, new structures appear[14,15] labeled 1, 1′ and 2, 2′ on Fig. 1.

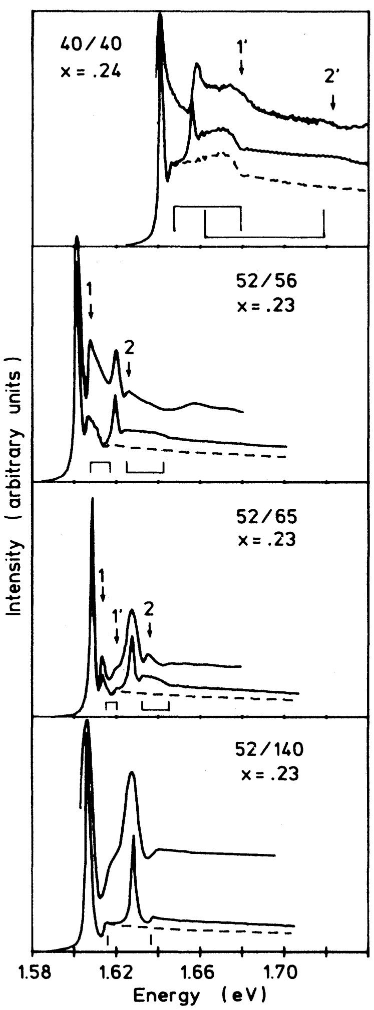


FIG. 1. Experimental excitation spectra (upper curves) and theoretical absorption spectra (lower solid curves) with the contribution of heavy-holes alone (dashed lines). The heavy-hole and light-hole optical transition energies corresponding to the center and to the edge of the Brillouin minizone are indicated below the curves. Sample parameters are given in the inset. Arrows 1, 1′ and 2, 2′, respectively, correspond to the $M_0, M_1$ singularities for HH and the $M_0, M_1$ singularities for LH.

We have reproduced the experimental results by computing the theoretical absorption spectra (Fig. 1, lower solid curves) with the method proposed in Ref. 7 (the calculation has been improved by including valence-band mixing effects), using the x-ray parameters of the real samples. Note the very close similarity between the experimental and the theoretical curves,[16] in particular as far as the shape and relative position of the new structures are concerned. The contributions of HH alone are represented by the dashed lines and the calculated $M_0$ (BZ center) and $M_1$ (BZ edge) optical transition energies for HH and LH are indicated in the figure. Theory clearly predicts that the existence of SL minibands is responsible for new structures in the absorption spectrum: structures 1 and 2 correspond to the $M_0$ singularities, and structures 1′ and 2′ correspond to the $M_1$ singularities. In the experimental spectra, all of these structures may not always be resolved because of the overlap of HH and LH contributions. Nevertheless, the sum of the electron and HH minibands can be directly estimated on the PLE spectrum in most cases, and also the sum of the electron and LH minibands in a few cases.

It should be stressed that the shoulder labeled 1′ on the (52 Å)/(65 Å) SL spectrum can no longer be attributed, as in the MQW case, to the onset of $M_0$ band-to-band transitions, since this would lead to a value of 13 meV for the binding energy of the $HH_0$ exciton, which is much higher than both experimental observations and theoretical estimates.[11] This structure is indeed related to the $M_1$ electron-HH singularity while the structure labeled 1 is related to the $M_0$ singularity, leading to the correct exciton binding energy and miniband width. When the SL period further decreases, the new structures 1 and 2 become broader and the shoulder 1′ disappears into the LH excitonic peak. Finally, in the spectrum of sample (40 Å)/(40 Å), both structures 1′ (HH related) and 2′ (LH related) can be resolved at high energies.

Structures 1 and 2, related to the $M_0$ singularity, are clearly enhanced in the experimental as well as in the theoretical spectra of the (52 Å)/(65 Å) and (52 Å)/(56 Å) SL's, compared to the case of MQW's. This might be indicative of a new excitonic character. In order to further investigate these structures, we have performed magneto-PLE experiments.

When a magnetic field is applied parallel to the growth axis of the superlattice, as in the case of MQW's, oscillations of the PLE intensity are observed. These oscillations can be plotted on a fan chart as Fig. 2 shows for the (52 Å)/(65 Å) SL. Let us first recall that in MQW's under magnetic field, the two excitonic transitions ($HH_0$ and $LH_0$) show a very small diamagnetic shift, and two series

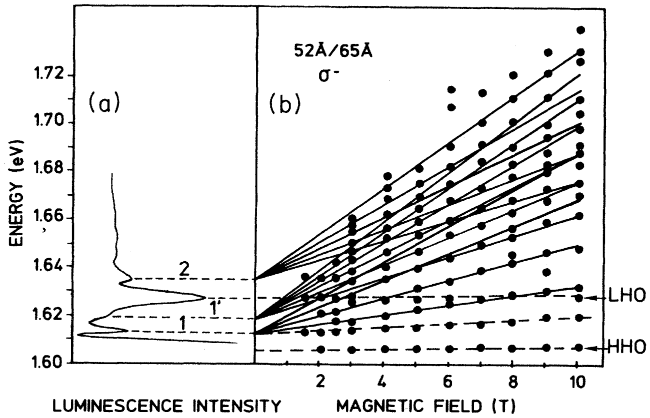


FIG. 2. Fan chart (right-hand side) and zero-field PLE spectrum (left-hand side) of the (52 Å)/(65 Å) sample. Lines are a guide for the eye.

of Landau levels are evidenced that approximately extrapolate to their respective band edges ($HH_0$ and $LH_0$) (see, for example, Ref. 17: see also Ref. 18 for theoretical discussion).

The following features can be observed on the series of SL's that we have studied.

(a) The two usual excitonic transitions $HH_0$ and $LH_0$ still show a weak diamagnetic shift.

(b) The binding energy of the $HH_0$ exciton is reduced: the first series of Landau transitions extrapolates to a point 5–6 meV above the $HH_0$ peak. This agrees with our present theoretical estimates as well as with previous calculations.[11] Taking into account this reduction of the exciton binding energy is necessary in order to interpret correctly the spectra.

(c) The new peak (labeled 1 in Fig. 1) also presents a weak diamagnetic shift and this shows the excitonic character of this transition. This confirms, as proposed by the theory, that new excitonic resonances are stable as a result of miniband dispersion in a SL.

(d) A new series of transitions is evidenced. This series corresponds to the new miniband dispersion in the SL. In a SL, when a magnetic field is applied parallel to the growth axis, the density of states is not a series of δ functions as in the case of a MQW, but is rather a series of bands with a sharp maximum at each edge.[19,20] Instead of one series of Landau levels for HH's we thus observe two series of transitions corresponding to the peaks in the density of states at $k_z=0$ and $k_z=\pi/d$.[21] In a way similar to the $k_z=0$ series which extrapolates to the $k_z=0$ band edge, the $k_z=\pi/d$ series extrapolates to the $\pi/d$ band edge. This confirms the assignments given by the theory.

Let us take the example of the (52 Å)/(65 Å) SL shown in Figs. 1 and 2: the structure 1′ at 1.62 eV, which would have been interpreted as the onset of the $k_z=0$ band-to-band transitions, is assigned, following the theory, to the onset of $k_z=\pi/d$ transitions. It does indeed correspond to the extrapolation point of the new series of transitions. In the same way, structure 1 is shown, following both theory and experiment, to be a superimposition of the onset of $k_z=0$ electron-HH band-to-band transitions (leading to the correct miniband width as well as exciton binding energy) and of a new excitonic structure arising from the SL miniband dispersion.

Let us note that the new Landau series is not at all related to the monolayer splitting[9] observed on the same samples for the following reasons: (i) we have detected the luminescence on the high-energy component of the luminescence, so that only one HH transition is observed in PLE at zero magnetic field. If we set the detection on the low-energy peak, all series of transitions are doubled due to this monolayer splitting. (ii) We also observe the new series of transitions on samples that do not show any monolayer splitting.

As a result of these observations, all the assignments given by our theoretical calculations are strongly confirmed. We can now state that new excitonic structures are indeed brought by the miniband dispersion and the existence of a saddle point in the superlattice dispersion relation. These resonances cannot be termed saddle-point excitons in the usual sense, as the miniband width is of the order of the binding energy of an exciton. The expansion of the exciton wave function thus cannot be limited to the neighborhood of the saddle point. The evolution of this resonance with the width of the SL minibands shows that no sharp resonance follows the $\pi/d$ extremum when the miniband width becomes large: see sample (40 Å)/(40 Å) in Fig. 1 (we have also studied SL's with periods down to 20 Å). If bound states exist, their lifetimes must be very short. Clear shoulders nevertheless appear in the theoretical and experimental spectra at the zone boundaries in $\pi/d$.

In summary, we have observed experimentally, studied in magnetic field, and fitted theoretically the new structures which appear in the photoluminescence excitation spectra of superlattices with electron miniband widths ranging from 0 to 30 meV. These new structures are due to the optical transitions at the center and at the edge of the Brillouin minizone in the growth direction, and need full treatment of the Coulomb interaction to be understood. Excitonic resonances are also found which might be the analog of saddle-point excitons in bulk materials. The agreement between theory and experiment is remarkable over the whole series of samples, and gives a very strong predictive value to the theory.

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